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16. Abstract

This synopsis discusses our development efforts in the area of resonant holographic interferometry (RHI). RHI is a completely nonintrusive optical diagnostic suitable for a wide range of gas dynamic investigations. As described in previous reports, RHI is a hybrid technique that combines the multi-dimensional imaging capability of holography, the phase sensitive detection of interferometry, and the species specificity of spectroscopy. When combined with tomography, RHI provides three-dimensional characterization and quantitative measurement of species concentrations, temperature, velocity, pressure, mixing and other thermophysical quantities.

Specific Aims - The specific aims of this program have not been modified from the original goals. These goals are to theoretically and experimentally demonstrate RHI by quantitatively measuring chemical species of interest to the Army in controlled combustion environments. Efforts this past year were focused on making quantitative RHI concentration measurements in a calibrated laboratory burner.

Results - During this reporting period RHI measurements of hydroxyl radicals (OH) were recorded in a Wolfhart-Parker (W-P) slot burner. The data is currently being analyzed and compared to absorption data and predictions based on our RHI model. The method of Phase Shifting Interferometry (PSI) was developed and implemented for converting an unwrapped phase map into concentration data. The PSI method was adapted for automation using a personal computer. A two-reference beam RHI system previously developed under this research program was modified to accommodate a calibrated W-P slot burner. Phase Shift Interferometry (PSI) data reduction was facilitated by using a Correcting Holographic Optical Element (CHOE) as described in previous reports and an automated computer data reduction package. RHI data at several burner flow rates were recorded using the RHI holocamera. The burner was calibrated using thermocouples and absorption measurements.

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STATEMENT OF THE PROBLEM STUDIED

The Army Research Office (ARO) supports research in combustion and propulsion aimed at its need for better performing propulsion systems. One goal of supporting such projects is an increased fundamental understanding of the storage, extraction, and subsequent conversion of chemical energy into work. The primary mode of energy conversion is combustion. Efforts to gain a more detailed understanding of the physics and chemistry of combustion are needed to increase the efficiency of these processes. These efforts take on the form of theory and experiment. Modeling is a more cost effective approach at improving combustor design and technology than empirical design; however experiments are still very much needed to validate the theoretical models, and to study complex combustors which cannot be analyzed with current models. Particularly useful parameters are temperature, species concentration, bulk flowfield density, and velocity. In addition, highly resolved spatial profiles of these parameters are sought for model validation.

Under sponsorship from the ARO, MetroLaser has developed the RHI technique to provide better measurements into the combustion process. RHI is a laser-based diagnostic that combines the imaging capability of <u>holography</u>, the phase sensitive detection of <u>interferometry</u>, and the species specificity of <u>resonance</u> spectroscopy. RHI provides a method of holographically recording species specific interferograms. RHI accomplishes this by recording two simultaneous holograms at two different laser wavelengths: one tuned near an absorption line, and the other tuned off this feature. Upon reconstruction, the resulting interference fringes correspond uniquely to the density of the species under interrogation because phase contributions from background species, thermal and pressure instabilities, and optical aberrations are common to both holograms and therefore are subtracted out in the reconstruction. This inherent background subtraction is one major benefit of recording holograms over standard interferograms.

This Final Report summarizes a program that represents the development and assessment of the quantitative nature of RHI measurements in combustion applications. Some of the attributes that make RHI attractive for combustion applications are:

- Operates on ground-state transitions (insensitive to quenching).
- Simultaneous imaging of gaseous species and solid or liquid object.
- Background-free "signals."

During the second three-year program we addressed the issue of quantitative measurement using RHI. Although RHI recordings can be easily inspected by eye to observe qualitative features of the combustion process, a quantitative measure requires computerized data reduction and sub-fringe resolution. Through the development and implementation of advanced signal processing and phase-shifting interferometry (PSI), we have established a procedure to make quantitative RHI recordings with a single laser shot. This higher degree of measurement resolution has allowed us to model the RHI process more accurately.

To better assess the quantitativeness of RHI, we designed our experiments around well-controlled, laboratory devices. The program began by performing species concentration measurements in a static cell of simple geometry. The target species was Na vapor. Temperature

control of the static cell ensured constant and known vapor pressures and consequently constant and known species number densities. The program then proceeded with measurements of OH in a well-characterized, steady-state flame. Comparisons were made against predictions, complimentary absorption measurements, and published data.

Although we had planned to also investigate tomographic inversion techniques to obtain the concentration of OH in non-symmetrical flames, the preceding comparison work between RHI measurements, absorption measurements, and predictions required more time than originally budgeted.

Phenomenology

The following phenomenological description was discussed in a previous report to the ARO; however, it is expanded upon and included here for completeness.[1]

RHI measures the phase changes experienced by a coherent laser beam as it propagates due to changes in the real part of the index-of-refraction of the medium. The resonant holographic interferograms are comprised of fringe patterns in which each fringe represents a contour of constant optical pathlength. These contours are then converted to species number densities. The real and imaginary parts of the complex refractive index are sketched as a function of wavelength in **Figure 1**. As illustrated, the index-of-refraction changes dramatically in the vicinity of a resonant absorption feature. Thus, by tuning one laser near the resonant feature and the other off resonance, a *species specific* hologram can be generated. The near-resonance laser beam is responsible for the fringes or contours of constant species density. The off-resonance laser beam accounts for all the species comprising the background. The off-resonance recording allows the background to be subtracted out in the reconstruction phase. Thus the resultant fringe pattern is attributable *only* to the species under interrogation.

An alternative approach used to increase RHI sensitivity by a factor of two is implemented by tuning each laser to corresponding maximum and minimum positions in the dispersion curve. Since the background contribution to the phase is a constant over this range, and the resonant contribution to phase is additive, the phase difference between the two waves is simply the sum of the absolute value of the phase shift due to the resonance effect. At the maximum and minimum positions, the absolute value of the phase shift is equal, and therefore the phase difference between the two waves is twice the phase shift due to any one wave.

In addition to phase information that results from changes to the real part of the index-of-refraction, RHI also stores absorption information that results from changes to the imaginary part of the index-of-refraction. This ability to store and measure multiple spectroscopic parameters is referred to as "multiplex spectroscopy". Another possible implementation of RHI is to access two separate transitions near-resonance. This provides information regarding the species' internal state population that allows the computation of temperature by assuming a Boltzmann distribution.
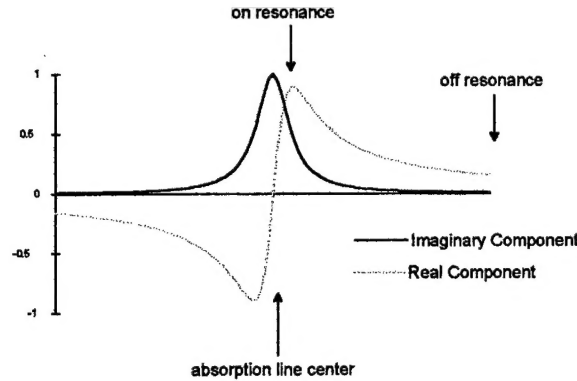
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Figure 1. Plot of Real and Imaginary components of the index-of-refraction near an absorption.

Approach

The objectives of the three-year program were to assess and demonstrate the quantitative capabilities of RHI concentration measurements. The approach consisted of modeling RHI measurement performance for a pair of target species, Na vapor and OH, and by experimentally verifying these predictions under controlled laboratory tests.

Analytical modeling was used to aid the RHI experimental designs, and verify the measurements. The model is capable of predicting phase changes, absorption losses, and laser-induced fluorescence resulting from the interaction between the laser field and the target species. The relationship between phase measurements and absorption-based measurements was established to provide a direct comparison between the calculated linear absorption and the RHI concentration measurements. The independent measurement provided the baseline calibration.

A master equation predicting RHI fringe shifts is written in a straightforward way. The pathlength through the target species can and does change across an RHI field-of-view, we simply assume the number density along any one of these paths to be uniform. We show later on that this assumption is not necessarily valid. The phase shift difference experienced by the two object beams is maximized by symmetrical tuning of the two lasers about the absorption feature at the half-maximum intensity positions.

The RHI fringe shift, F, is given by the integrated phase difference between the two recorded holograms divided by 2π. We write it as

$$F = 2 K f L N \frac{g_{dis}^*(\omega_o, \omega_l)}{2\pi} \quad . \tag{1}$$

The object beams that generate the RHI hologram also experience absorption. From Beer's law we can write an equation similar to the one above which expresses the transmission, T, of the object beams through the test volume as

$$T = \exp\left(-4\pi K f L N g_{abs}^*(\omega_o, \omega_l)\right) \quad . \tag{2}$$

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In the above, we have used the following symbols:

K a constant (2.24×10^{-14} cm).
f species oscillator strength (dimensionless).
L pathlength through target species (cm).
N target species number density along path L (cm^{-3}).

The functions $g^*_{dis}(\omega_o, \omega_l)$ and $g^*_{abs}(\omega_o, \omega_l)$ (units of cm) denote the weighted, normalized, real and imaginary parts of the refractive index of the target species. These two functions are weighted by the normalized frequency distribution of the laser, $g_L(\omega-\omega_l)$, as follows,

$$g^*_i(\omega_o, \omega_l) = \int g_i(\omega - \omega_o) g_L(\omega - \omega_l) d\omega \quad . \tag{3}$$

The resonance frequency of the target species is denoted by ω_o. The central frequency of the laser by ω_l. For most applications, $g_{abs}(\omega-\omega_o)$ is given by the Voigt function and $g_{dis}(\omega-\omega_o)$ by its antisymmetric counterpart.

The controlled laboratory testing environments included a static sample cell and a Wolfhardt-Parker slot burner, for the Na vapor and OH work, respectively. The choice of a static cell design was predicated on the need to first establish the viability of previously developed data reduction procedures, and put them into practice for the first time. A necessary initial step is choosing conditions of simple geometry that will allow a straight-forward inversion of the phase maps into absolute species number density profiles. The simplest system is a static sample cell. The sample cell was evacuated and a small amount of an alkali metal was placed inside. The cell was then inserted into a temperature controlled tube furnace. The tube furnace allowed precise control of the vapor pressure, and thus the amount of alkali metal in the gas phase. For constant volume conditions, the vapor pressure is directly proportional to the temperature of the cell, and therefore, a precise amount of alkali metal was introduced into the object path of the RHI spectrometer. The sample cell itself has a wedged-shape that offers variable pathlength over the field-of-view.

The choice of alkali metals as the target species for the static sample test cell experiments was based on several reasons. First, MetroLaser had already successfully made RHI measurements of both Na and K. Second, alkali metals exhibit strong, well-isolated, and well-characterized spectral features that are easily reached with the lasers available to this program; and third, there is much interest in the use of alkali metals as seed species in combustion, plasma, and shock diagnostics.

As the next logical step in our progression to demonstrate the quantitative capabilities of RHI measurements, we moved the program from static test cells to well-characterized laboratory burners. The design of experiments to move the program in this direction centered on the selection of a well-characterized, benchmark laboratory burner. Not only must the combustion device behave predictably, but it must stand as a widely accepted standard within the combustion community. Only then will a quantitative, RHI measurement demonstration withstand the scrutiny of the peer review process and ultimately become accepted by practicing combustion engineers.

We chose to perform the RHI development work on a standard Wolfhardt-Parker (W-P) burner. The W-P burner is a slot diffusion flame device. The fuel flows through a rectangular slot. Air flows along the perimeter of the slot. Unlike the "flat flame" profiles of the McKenna and Hencken burners, the W-P burner exhibits an inverted "V" shaped flame. The flame is one-dimensional along the slot. Flame temperatures are on the order of 2300 K. We calibrated the W-P burner using a radiation corrected, R-type thermocouple.

The target specie for this second phase of RHI experiments was the OH molecule. We have previously recorded high quality RHI holographic interferogram of OH in several combustion environments (Bunsen burners, slot burner, and shock heated flows). The spectroscopy of the OH molecule is well-understood, and OH is a ubiquitous species in combustion reactions.

An independent means of calibrating the flow was also established. For this task we adopted linear absorption. Several absorption measurements within the RHI field-of-view anchored the RHI holographic interferogram to an absolute OH number density.

For the ARO development work, the RHI instrument employed two, independently tunable, narrow-band, short pulse laser sources. The independent tunability gave the maximum flexibility in selecting resonance transitions, and in setting the on- and off-resonance wavelengths. This also allows access to two separate transitions for the possibility of measuring temperature. The narrow band feature has two benefits. The first is the increase in instrument sensitivity with decreasing laser linewidth. The second is the concomitant increase in the beam coherence length. Longer coherence lengths facilitate the matching of the object and reference beam path lengths, a requirement for high quality interferograms. The short pulse feature provides excellent time resolution for investigating rapidly fluctuating flows.

The schematic of a typical RHI breadboard used for many of our experiments is shown in Figure 2. For the simultaneous recording of a two-wavelength resonance hologram, the timing of the on-resonance λ_1 and off-resonance λ_2 beams must be overlapped within the appropriate time scale for the phenomenon under investigation. For example, some turbulent features exhibit fluctuations on the order of 10 kHz or higher. A temporal separation of better than 100 ms. Our pulse separation was typically set at 10 – 20 ns. The motion freezing capability of pulsed experiments is critical when probing turbulent flow fields, as exist in many plasma and combustion situations. The temporal overlap is accomplished through optical delay lines. The temporally overlapped on-resonance and off-resonance laser beams are collinearly combined to form the object beam. The object beam, comprised of both wavelengths, is directed through the probed medium. In this research the probed medium was either contained inside a controlled sample cell or generated with a calibrated burner. The residual laser light is used for the two independent reference beams. Each reference beam is comprised of a single wavelength of light. This is necessary to implement the data reduction method of phase shifting interferometry (PSI). The two reference beams traverse a distance equal to that of the object beam, bypassing the probe volume. The object beam and the reference beams are directed to a holographic film plate where they interfere to produce the resonance hologram. Pulse energy requirements to expose a 50 mm diameter hologram (Kodak 120, 125, or Agfa Gavaert 8E56) are on the order of 5 mJ in an 8 ns pulse.[2,3,4] These requirements are easily met with commercial pulsed dye lasers.
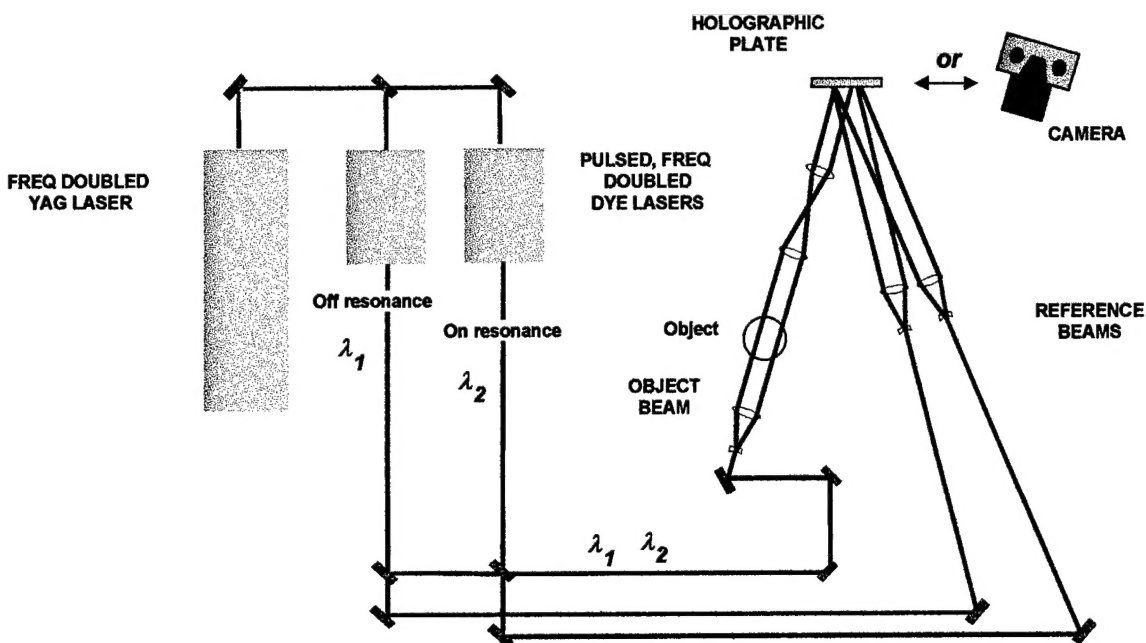
Figure 2. Schematic diagram of RHI instrument. Holocamera was replaced with 35 mm camera for transmission imaging measurements. In this study, the object was either a static sample cell or a calibrated slot burner. The two reference beams are necessary for the implementation of Phase Shifting Interferometry.

RESULTS

Static Test Cell

The data associated with Na vapor in the wedge-shaped static sample cell is depicted in Figure 3. The panels in the figure show an end-on view. In the region where the object laser beams do not interact with the sample cell, a series of evenly spaced reference, or tilt, fringes will be observed. The spacing of the reference fringes will be a function of colinearity of the two beams making up the object leg of the RHI spectrometer and the wavelength difference between the two beams. Both colinearity and wavelength difference are controllable experimental parameters. The phase-shifted data fringes appear only in the region where the object beams interact with the contents in the sample cell. The spacing and periodicity of the data fringes are shifted due to the number density of the targeted species, the pathlength difference across the field-of-view, and the oscillator strength of the transition probed.
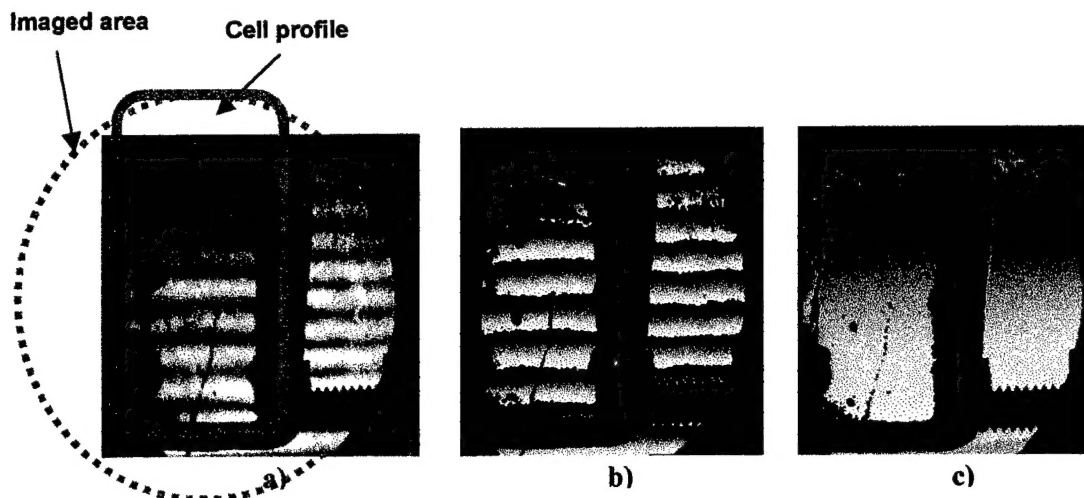
Figure 3. End-on view of RHI images of Na vapor in the static test cell. The raw RHI finite fringe interferogram is displayed in panel (a). The corresponding wrapped phase map is shown in panel (b). Panel (c) is the unwrapped phase map uncorrected for tilt.

Slot Burner

The data associated with OH measurements in the W-P slot burner is depicted in Figure 4. Corresponding line-of-sight absorption measurements were taken at four heights above the burner surface. The RHI image is an infinite fringe interferogram, thus the visible fringes correspond to regions of high OH number density. The RHI and absorption images were recorded on different shots during the same day.
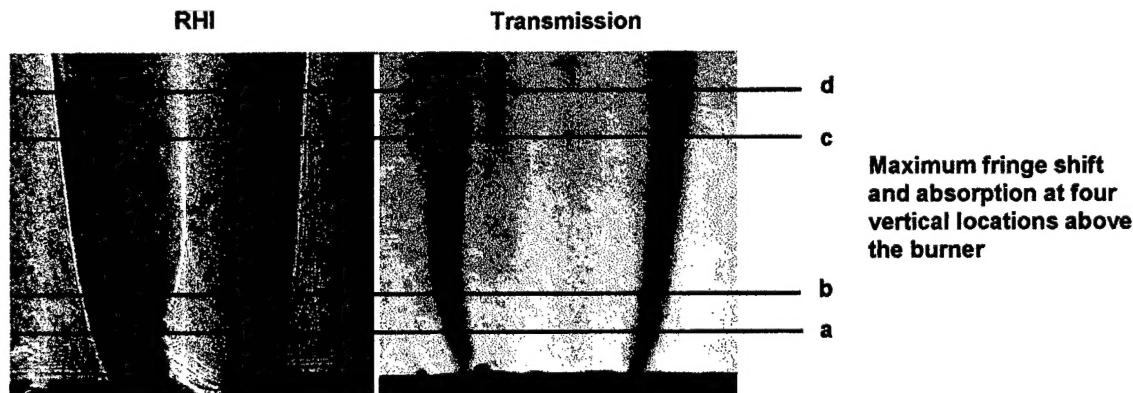

Figure 4. RHI and transmission data of OH generated in the Wolfhardt-Parker slot burner.

ANALYSIS

The applicability of holographic interferometric techniques for combustion measurements depends on change in flow density or species within the flowfield. The effect of an altered density field (and the resulting change in index-of-refraction) alters the optical phase between two wavefronts that pass through the field. The result is an interferogram of sinusoidal fringe

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patterns that represents the changes in the optical phase density (OPD) as light rays sample the flowfield.

Each light ray that passes through the test section experiences a propagation delay, depending upon the local refractive index (n) at each point along the path of the ray. The refractive index has a unique relationship with density (ρ), and is represented as $n-1=\rho/K$, where K is the Gladstone-Dale constant. If the boundary conditions and optical alignments stay constant, the change in optical phase distribution $\Delta\phi(x,y)$ between two exposures can be written as

$$\Delta\phi = \frac{2\pi}{\lambda} \int_{path} \Delta n dl \quad , \tag{4}$$

where Δn is the change of refractive index distribution in the test section, λ is the wavelength, and l is the optical path-length inside the test section.. Therefore, an interferogram is the combination of many point functions $\Delta\phi(x,y)$, which form the fringe pattern. The map of dark and light "fringes" represents the phase delay of the light passing through the test section. Each individual fringe order (either light or dark) marks a specific increment of phase change relative to the adjacent fringe order. The results of a fringe analysis (given certain assumptions about the symmetry of the flow) produce a map of refractive index distributions, which represents the density distribution in the flowfield. The distribution may be calibrated absolutely either through the application of the model or complimentary measurement using linear absorption.

The intensity of the interferogram at any one point (x,y) is given by the following formula:

$$I(x,y) = A + B\cos\left(\frac{2\pi\cdot OPD(x,y)}{\lambda} + \omega\right) , \tag{5}$$

where A and B are constants related to the amplitudes of the two plane waves, OPD represents the integrated path-length difference, and ω is a constant phase angle dependent on the experimental setup. In areas where the OPD is not constant, fringes result, as described by the cosine term of the above formula.

For molecular target species with resonant oscillator strengths much less than unity, the approach to modeling is straightforward. Under such conditions we can safely assume that the incident electromagnetic fields do not perturb the local refractive index and that the molecular response does not perturb the incident fields. This paradigm permits us to calculate the phase shift due to a single molecule and sum the result over N such molecules presumed to be evenly distributed along the optical path through the target zone. The molecule is viewed in this scenario as a linear harmonic dipole responding to the incident field. Expressions for the contribution to the complex refractive of such a simple oscillator are readily found in textbooks. Typically, such discussions restrict themselves to the case in which the absorptive frequency response is given by a simple Lorentzian line shape. This is fine if Doppler broadening can be ignored with respect to natural lifetime or pressure broadening. For the OH molecules in our test environments, the Doppler and pressure broadening are of the same magnitude. Therefore, we

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When using the above model to predict or analyze measurements, three input parameters are needed: a) the oscillator strength of the molecular resonance, b) an estimate of the Doppler width (found from an estimate of the temperature), and c) an estimate of the pressure broadened linewidth. (We assume here that the target species concentration is being used as the dependent or fit variable in comparison to measurements.) The uncertainty in the calculated RHI fringe shift is linearly related to the uncertainty in the oscillator strength value used. For OH this uncertainty amounts to ±20%.[5] Hence, the calculated fringe shift has an uncertainty of ±20% associated with it. The Doppler width varies as the square root of the temperature. Hence, the associated uncertainty is typically smaller than that of the oscillator strength. The affect of the uncertainty in the pressure-broadened width on the estimated fringe shift must be evaluated on a case–by–case basis. Its influence will depend upon where the lasers are tuned with respect to line center and the ratio of the Doppler to pressure-broadened contributions to the linewidth. Figure 5 below helps to explain this. The red curve traces out the dispersive OH response for the conditions of the flame studied here. The blue curve traces out the response for the same flame at twice the pressure. Note that near the two extrema points, the response changes very little while out in the wings the change is more significant. The measurements reported here used lasers tuned near the extrema. This reduces the sensitivity to uncertainty in the pressure-broadened component of the linewidth.
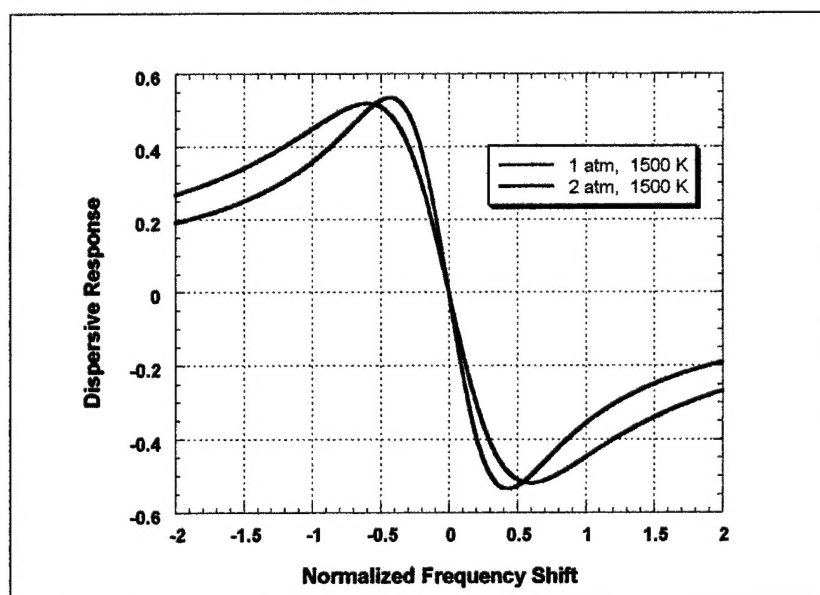

Figure 5. Dispersive response for OH at two flame conditions: a) 1 atm, 1500 K and b) 2 atm, 1500 K. The Doppler width (0.1 cm^{-1}) is the same for both while the pressure broadening contribution to the linewidth changes by a factor of two: a) 0.1 cm^{-1} and b) 0.2 cm^{-1}.

Despite its simplicity the above model does a reasonable job of predicting experimental outcomes. This is evident in the RHI data for OH shown in Figure 6. The points labeled **a – d** correspond to the vertical positions within the flame as previously indicated in Figure 4. The model agrees with the experiment to within a factor of two for this data. The larger deviation for the upstream points (a-c) is most likely a function of spatial and temporal resolution. The reaction zone within the flame is much tighter and well-defined at positions closer to the burner

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surface. At downstream positions, the reaction zone becomes more diffuse. Therefore, as the flame wafts from side to side on any given laser shot, the reaction zone is more likely to move out of the laser probe volume for upstream positions. The more diffuse flame at downstream positions is more likely to remain within the laser probe volume.
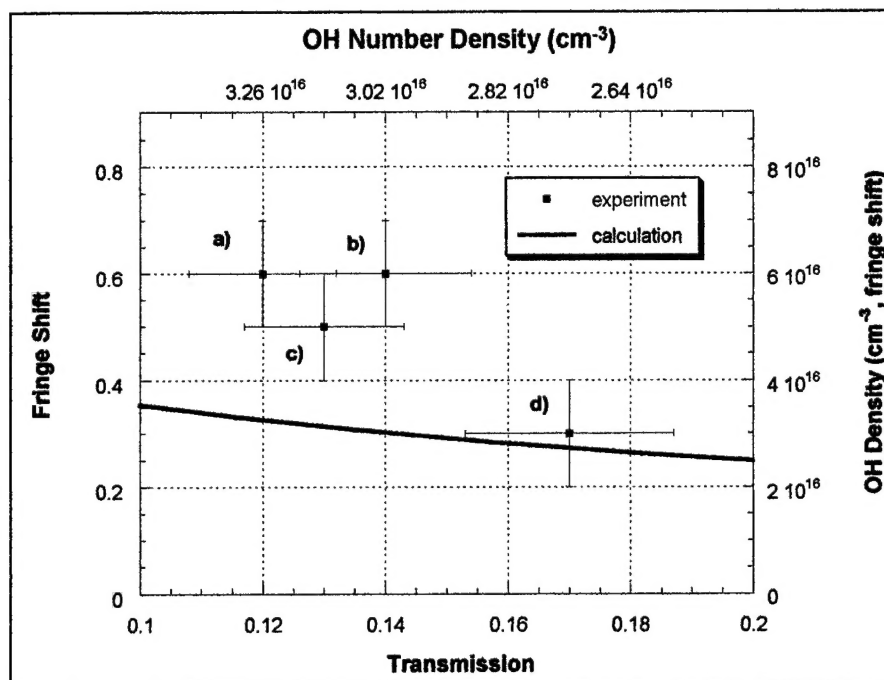

Figure 6. RHI data for OH in the Wolfhardt-Parker slot burner. Labels a-d correspond to vertical positions above the burner surface.

We note that during the execution of this program we recognized that at the detail level some texts are amiss with factors of pi when discussing the frequency response of the refractive index. Typos of this nature make direct numerical comparison between transmission and fringe shift impossible if they are not corrected. In particular, practitioners are advised to make sure that their expressions for the absorptive and dispersive line shapes are self-consistent with regards to constant scaling factors.

Numerically, estimating a RHI fringe shift comes down to taking the difference between two numbers, $k_1 n_1$ and $k_2 n_2$ where k_1 denotes the wavevector of laser beam number one and n_1 the refractive index at that same frequency (similarly for k_2 and n_2). Since the wavevectors differ by a small amount it is tempting to set them equal to each other when cranking out numbers. In practice this can lead to erroneous results and we caution against it.

For atomic species with oscillator strengths near unity, the simple model becomes inadequate. For sufficiently high densities and/or incident field strengths, the fields and atomic response become coupled, making the non-perturbative assumption invalid. In such cases, one would need to use a rigorous radiation transfer approach to model the measurements. This became evident upon our examination of the Na vapor data. Although, as shown in Figure 7, the RHI data for Na vapor exhibited smooth, linear behavior, this was often misleading as we found that the model

under predicted the observed fringe shift. To compensate for this discrepancy, additional modeling efforts will be required. The modifications to the model will most likely involve the inclusion of pulse propagation effects.
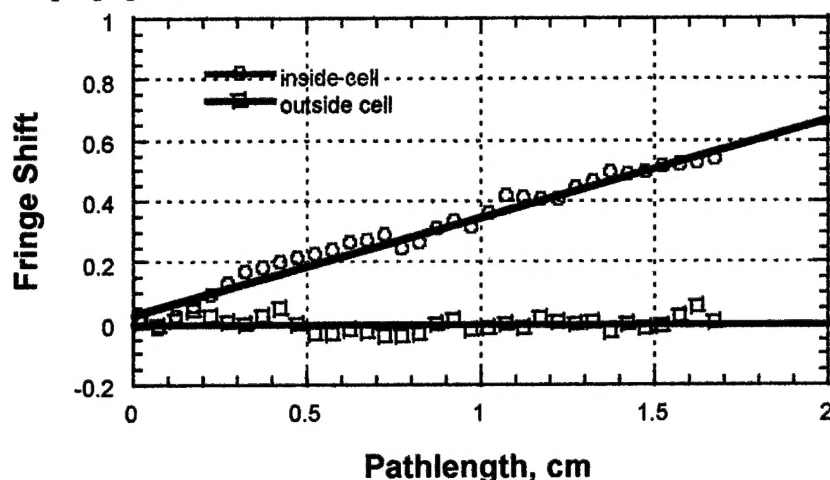

Figure 7. Linear response of the Na vapor RHI data.

SUMMARY

We addressed the issue of quantitative measurement using RHI applied to combustion measurement problems. The assessment required the development of models and the performance of experiments. Comparisons were made against predictions, complimentary absorption measurements, and published data. For probing environments of moderate optical densities, such as molecular species with moderate oscillator strengths far from saturation conditions, our model satisfactorily predicts the RHI measurement. For environments exhibiting high optical densities, such as atomic species with close to unit oscillator strength near field intensity saturation conditions, the present model is inadequate.

In terms of future RHI development work, there are two areas requiring additional investigation. To account for measurement discrepancies under saturation and/or high optical densities, additional modeling to account for pulse propagation effects will be needed. To increase the practical utility of the RHI technique, investigations of tomographic methods for full field measurements are recommended.

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